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<b>14. ABSTRACT</b> ThermoDynamic Films LLC (TDF), in collaboration with the University of New Mexico (UNM), improved the performance of optical refrigerators for deployment space-borne missions and terrestrial applications. They have optimized the ytterbium doping concentrations the optical refrigerator cooling elements. With these advances the TDF/UNM collaboration has been able to an optical-refrigerator cooling element comprised of an ytterbium-doped yttrium lithium fluoride (Yb:YLF) crystal from room temperature to 93 K. This is the world record for optical refrigeration (or for any solid-state cooling technology). They have developed an externally pumped optical cryocooler device, which enhances the energy density of laser light in the cooling cavity to increase the efficiency of converting laser power into cooling power. They designed and built high thermal conductivity links that connect cooling crystal to the load while providing shielding of the cold elements from the intense fluorescence generated in the cooling process. They also developed an optical refrigeration scheme based on intracavity-enhanced absorption that uses optically pumped surface-emitting lasers.					
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**Final Report for  
STTR Phase II: Optical Refrigeration for Dramatically  
Improved Cryogenic Technology**

Contract number: FA9550-13-C-0006

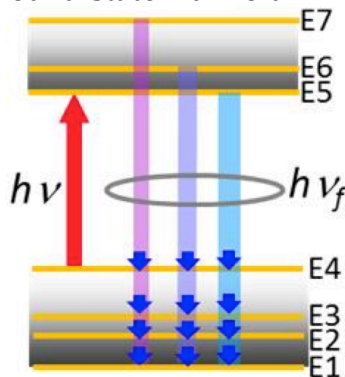
Contract line item number: 0001AG

Performance period: 1 July 2014 through 31 October 2014

**Background:**

Optical refrigeration or laser cooling of solids is the subject of intense current interest because it is the only proven mechanism for all-solid-state, high performance cryocooling [BCE98, RIE09, DVS12]. This rapidly emerging technology offers the advantages of compactness, high reliability, no vibrations, no moving parts, no electromagnetic interference and no need for cryogenic working fluids. The low-temperature performance of optical cryocoolers far surpasses that of thermoelectric coolers, the primary competing solid-state refrigeration technology. The TDF/UNM collaboration has pioneered this field and has continued to be its dominant leaders.

The most successful optical refrigerators are based on ytterbium-dope crystals, ytterbium-doped YLF crystals (Yb:YLF) in particular. Figure 1 is a sketch of the low-lying energy levels for ytterbium-doped crystals. The ground state and first excited state of the ions are split by crystal-field interactions. To cool these materials, one illuminates them with laser light at frequency  $h\nu$  that is tuned to the energy difference between the top of the ground-state manifold and the bottom of the excited-state manifold; this corresponds to a wavelength of 1020 nm for Yb:YLF. The ions absorb the light and jump to the bottom of the excited manifold. The excited ions then absorb thermal energy from the solid before radiatively decaying to the ground-state manifold levels by emitting photons with average fluorescent energy  $h\nu_f$  that is greater than the energy of the pump photon. The cooling cycle is completed when the ions absorb more thermal energy from the solid to repopulate the top level of the ground-state manifold.



**Figure 1.** The cooling cycle for ytterbium-doped solids. This is not to scale. The actual energy scale is  $h\nu_f \sim 1.24$  eV, while the width of the ground-state manifold is only about 0.05 eV.

For the optical cooling cycle to operate, there has to be sufficient population of thermally excited ions near the top of the ground-state manifold to absorb the laser light. This condition implies that the width of the ground-state manifold is not too much greater than the thermal energy  $k_B T$ , where  $k_B$  is Boltzmann's constant. As the temperature falls,  $k_B T$  eventually becomes much smaller than the energy width of the ground-state manifold, and the population of ions near the top of the ground-state manifold decreases so much that it becomes difficult to cool further.

The cooling efficiency  $\eta_c$ , defined as ratio of the cooling power to the absorbed power, is given by [MSB07],

$$\eta_c = \eta_{ext} \eta_{abs} \frac{h\nu_f}{h\nu} - 1 \quad (1)$$

where the product of external quantum efficiency ( $\eta_{ext}$ ) and pump absorption efficiency ( $\eta_{abs}$ ) describes the probability of conversion of an absorbed photon at  $h\nu$  to a fluorescence photon with mean energy  $h\nu_f$  exiting the material. The factor  $\eta_{abs}$  accounts for loss of pump light to parasitic background absorption, and the former factor  $\eta_{ext}$  describes reduction of cooling efficiency due to: i) luminescence trapping in the sample and ii) optical excitations that recombine non-radiatively. When these efficiency coefficients are significantly less than unity, undesired heating will occur. Understanding, quantifying, and addressing these two efficiencies ( $\eta_{ext}$  and  $\eta_{abs}$ ) has been the key to attaining efficient optical cryocooling.

In 1995, net laser cooling in solids was first achieved at Los Alamos National Laboratory (LANL) by a team led by TDF's CEO [RIE95]. Two technical challenges were addressed and overcome in these experiments. A material system was needed in which i) the vast majority of optical excitations recombine radiatively and ii) there is minimal parasitic heating due to unwanted impurities. It was also important that spontaneously emitted photons escape the material without being trapped and re-absorbed, which would effectively inhibit spontaneous emission. This is a critical issue for cooling high-refractive-index semiconductors such as GaAs, where total internal reflection can cause strong radiation trapping [MSB07]. The efficiency of optical cryocoolers can be improved by converting the waste fluorescence to electrical energy via strategically placed photocells; such a device can approach the Carnot efficiency limit [MSB09].

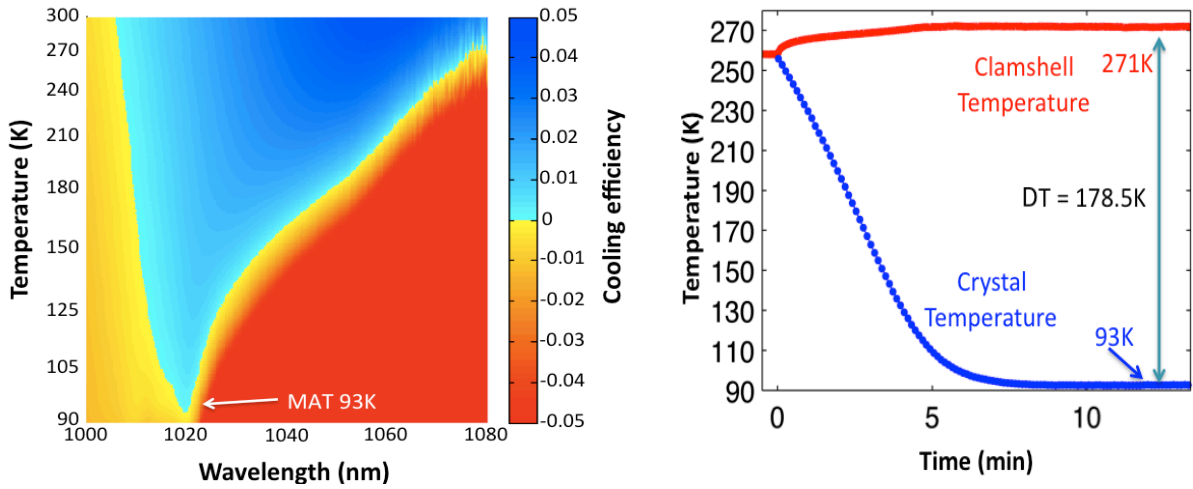
In 2009, the UNM group achieved record cooling with an 5% Yb-doped YLF crystal grown at the University of Pisa, Italy [DVS10]; the Yb:YLF crystal cooled by 150 K with respect to the absorbing chamber (which is referred to as a "clamshell") and reached an absolute temperature of 155 K. The cooling power (*i.e.*, heat lift) was over 100 mW at the lowest temperatures attained at that time. To put this result in perspective, all-solid state commercial thermoelectric coolers based on the Peltier effect require multiple stages to approach an absolute temperature of 170K with heat lift of < 50 mW.

## Phase II accomplishments:

To effectively cool Yb:YLF crystals UNM acquired an ytterbium fiber laser system that operates at maximum of 60 W at  $\lambda \sim 1020$  nm. With this laser the UNM/TDF team was able to set new performance milestones in laser cooling of solids and brought optical refrigeration into cryogenic temperature regime as defined by the National Institute of Standards and Technology ( $<123$  K). During this Phase II, even lower temperatures were achieved: 93K [MEL14]; see Fig. 2.

The limits of cooling for a specific cooling element can be understood through analysis of separate experiments to ascertain the minimum achievable temperature (MAT). The cooling efficiency described by Eq. (1) depends on wavelength and temperature as follows:

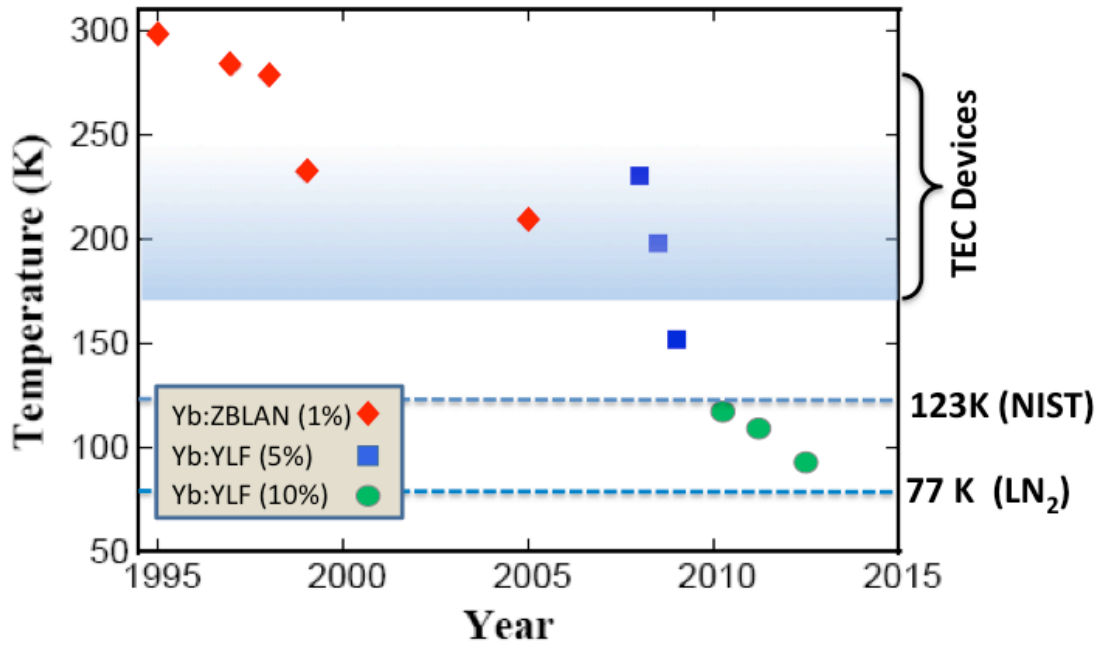
$$-\eta_c(\lambda, T) = 1 - \frac{\eta_{ext}(T)}{1 + \alpha_b / \alpha_r(\lambda, T)} \times \frac{\lambda}{\lambda_f(T)} \quad (2)$$



**Figure 2.** (Left panel) “Cooling map” for a 10% doped Yb:YLF crystal as a function of pump wavelength and temperature from Eq. (2). Blue indicates regions where cooling occurs and red corresponds to heating. (Right panel) The measured change in temperature with time [MEL14].

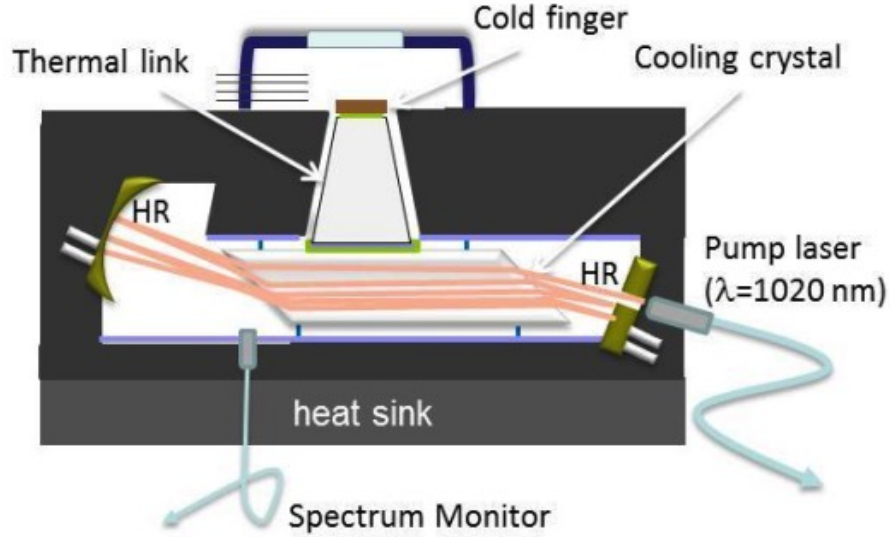
Temperature-dependent fluorescence spectra provide the mean fluorescence wavelength  $\lambda_F(T)$ ; other measurements gives the temperature-dependent resonant absorption  $\alpha_r(\lambda, T)$ . Over the range of pump wavelengths ( $\lambda$ ) the background absorption can be taken as constant. For the sample described in Figure 2, the value of the background absorption is found to be  $\alpha_b = 4 \times 10^{-4} \text{ cm}^{-1}$ . The resulting “cooling map” (a contour plot of cooling efficiency versus the temperature of the material and the wavelength of the laser light) shows that the minimum achievable temperature (MAT) is 93K. The data shown in the right panel of Figure 2 shows the cooling when the Yb:YLF crystal is pumped at the  $E4 \rightarrow E5$  transition of  $\text{Yb}^{3+}$  at 1020 nm. The experimentally achieved MAT was 93, in agreement with predictions.

The recent milestone achieving cryogenic optical refrigeration can be better appreciated by examining the record of minimum temperature achieved versus year since the realization of optical refrigeration in 1995 (Fig. 3). The current record low temperature of 93K is with a crystal with  $\alpha_b = 4 \times 10^{-4} \text{ cm}^{-1}$ . With further purification to reduce  $\alpha_b$  to  $1 \times 10^{-4} \text{ cm}^{-1}$  and/or increasing the  $\text{Yb}^{3+}$  doping concentration, cooling down to 70 K with Yb:YLF is within reach.



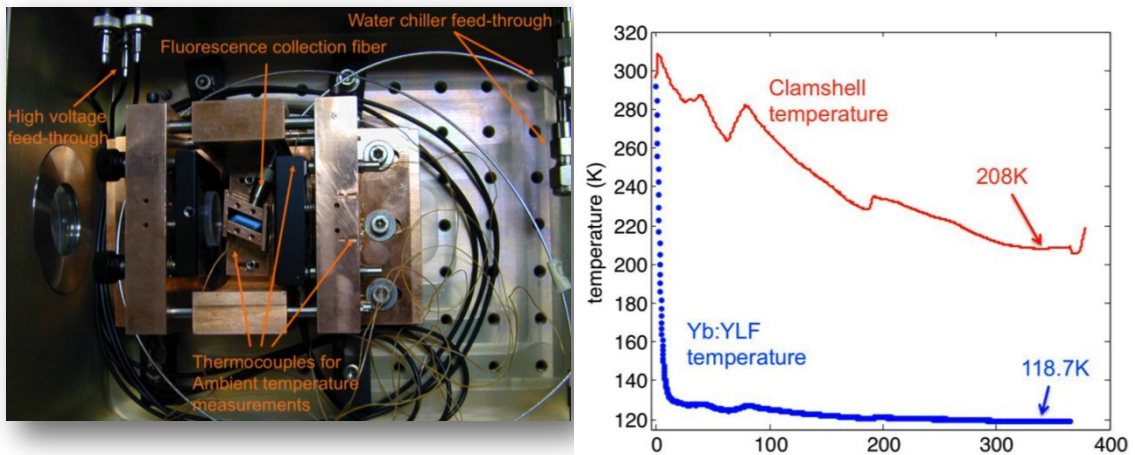
**Figure 3.** Record minimum temperature has decreased since the first demonstration of laser cooling of fluoride glass at LANL in 1995. In 2009, optical refrigerators cooled below 170 K, the lowest temperature accessible with current thermoelectric coolers. The “NIST cryogenic barrier” at 123 was surpassed soon after. Recent experiments have shown cooling at 93K. The next big goal is to reach liquid nitrogen temperature at 77K.

To demonstrate low-temperature optical refrigeration The TDF/UNM team developed Externally-Pumped Optical Cryo-Cooler (EPOCC) devices, as shown schematically in Fig. 4 and in a photo in Fig 5. In this device the energy density of laser light in the cooling cavity is enhanced through a multipass nonresonant scheme. The external laser can be either a fiber laser (Yb-fiber) or a diode laser operating at 1020 nm. In these experiments, a fiber laser was used. The pump laser is coupled through a hole in the front dielectric mirror and is carefully aligned to obtain 6-8 round trips. This approach cooled a 5% doped YLF crystal to 118K [SDM12].



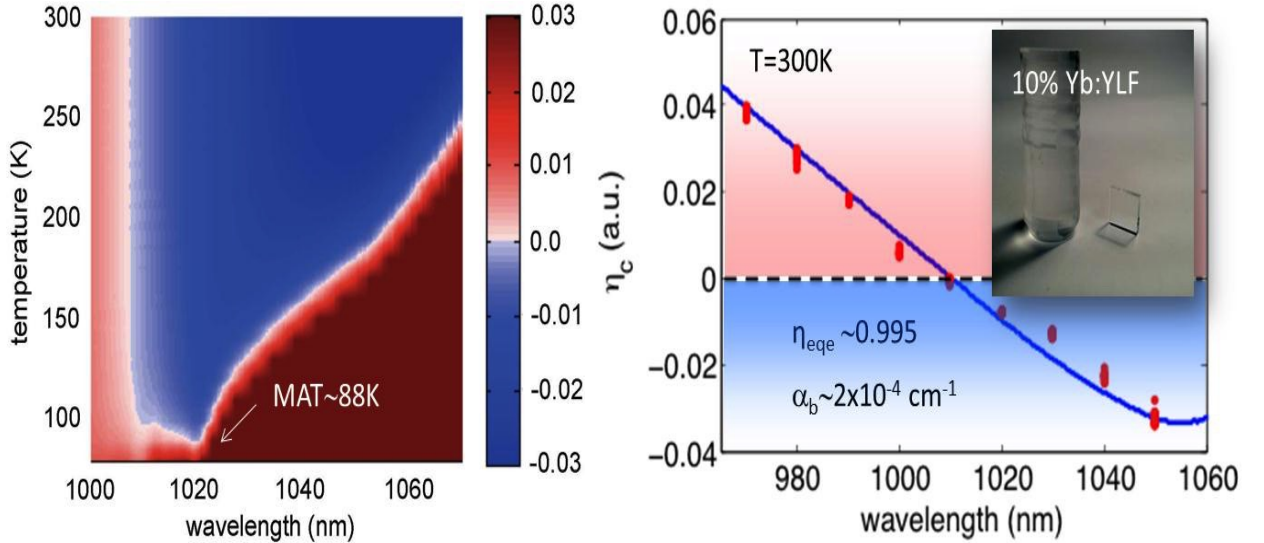
**Figure 4.** Schematic of an Externally Pumped Optical Cryo-Cooler (EPOCC) using a Yb:YLF crystal. The laser light at 1020 nm is carried by a fiber and coupled through a hole in the entry mirror.

A systematic optimization of dopant (Yb) concentration in YLF crystals is essential for creating optical cryocoolers that operate at the lowest temperatures with the highest efficiencies. The TDF/UNM team collaborated with Dr. Hans Jensen (AC Materials, FL) for growing high purity Yb:YLF crystals with various doping levels. Dr. Jensen produced crystals with doping concentration of 1%, 5%, 7% and 10%. All samples were screened using UNM's test facilities and were found to be "cooling grade according to Eq. 2.



**Figure 5.** (Left panel) A photo of the experimental setup employing EPOCC absorption arrangement inside a vacuum box. (Right panel) cooling results showing the temperature of the Yb:YLF (blue line) and clamshell (brown line) as a function of time. The sample temperature reaches 118.7K approaching the predicted MAT (110K) of this sample.

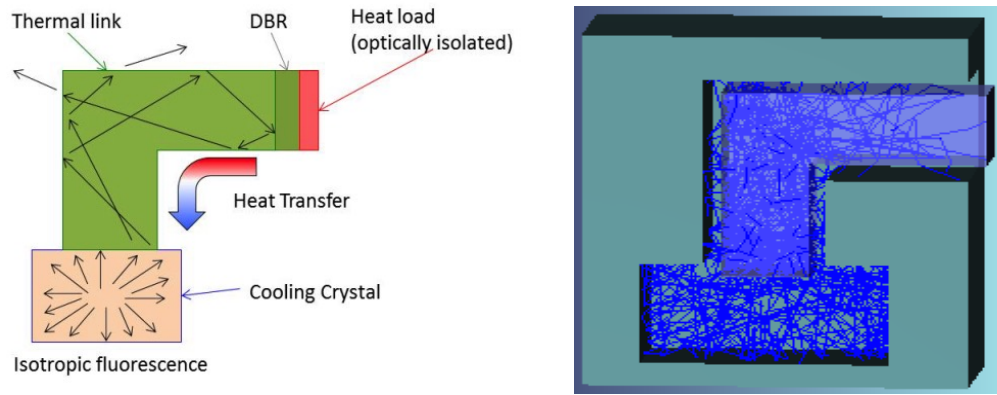
In particular, the 10% doped crystal has performed well with a predicted a minimum achievable temperature *MAT* of 88K. Preliminary room temperature data are shown in Fig. 5 (right), where measured temperature change versus laser wavelength is shown. Data analysis based on the standard laser-cooling model indicates  $\eta_{\text{ext}} \sim 99.5\%$  and a background absorption coefficient  $\alpha_b \sim 2 \times 10^{-4} \text{ cm}^{-1}$ . These remarkable characteristics, in turn, lead to a calculated “cooling map” (Fig. 6. Left) which predicts a *MAT* of 88K. Since the first observation of cryogenic operation in the 5% Yb:YLF, this presents a significant milestone in the optical refrigeration technology; a substantial progress towards development of a practical high efficiency and compact all-solid-state cryocooler operating liquid nitrogen temperature (77K) and beyond. Equally notable is the fact that this work relies on a domestic collaboration (AC Materials, FL) for this highly promising material synthesis.



**Figure 6.** (Left panel) Calculated cooling efficiency map as a function of temperature and laser wavelength for the measure parameters of the 10% doped Yb:YLF crystal. A *MAT* of 88K is predicted. (Right panel) Room temperature measurements that determine the crystal parameters (the inset shows the cooling crystal and the boule from which it was cut).

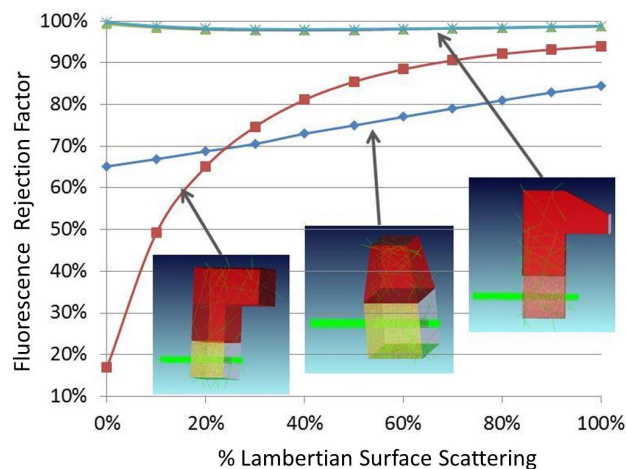
The cooling element of an optical refrigerator cools nearly uniformly as heat is removed by the luminescence. To cool a load, such as an infrared detector, the cooling element has to be linked to the load, so it can draw heat from it (Fig. 7). This coupling sets up a temperature gradient between the element and the load. In a good thermal link, this temperature gradient is minimized. At the same time, very little of the fluorescence can be allowed to impinge on any parts of the load; otherwise, radiative heating could easily overwhelm the optical refrigeration effect.





**Figure 7.** Kink-style thermal link. Left: Heat from the load readily diffuses around the kink. The luminescence largely escapes from the sides of the link when it encounters the sharp angle. Right: A ZEMAX simulation of the light shedding.

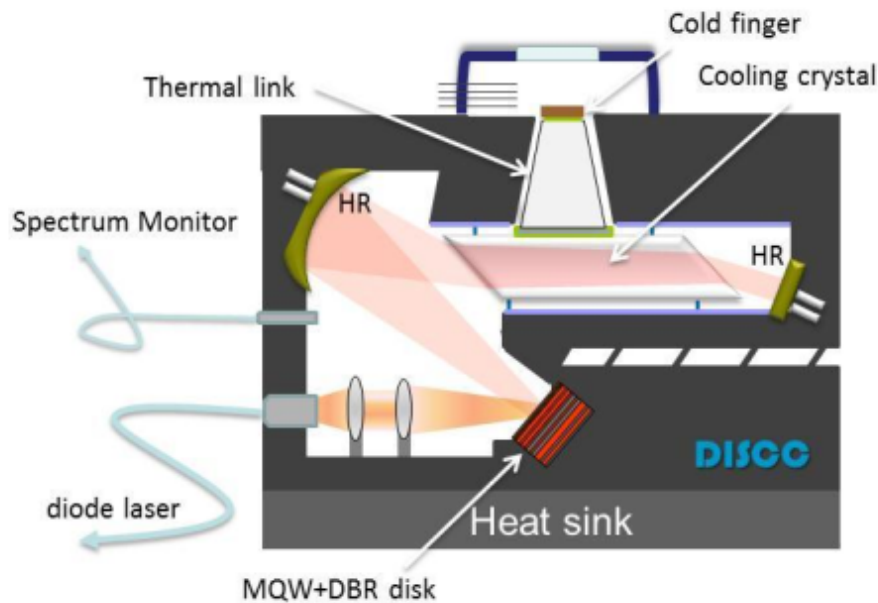
A practical thermal link in optical refrigerators has always posed an engineering challenge: it requires a high thermal conductivity interface while providing shielding of the load on the cold finger from the fluorescence. Nearly all designs that have been studied with modeling and experiments have relied on the use of dielectric Bragg mirrors which introduce undesired thermal resistance and some degree of heating. The TDF/UNM team considered novel thermal links that do not use any Bragg (dielectric or semiconductor stacks) mirrors. A number of designs including tapered-kink thermal link (TKTL) geometries were considered and modeled using ray tracing software ZEMAX. The results for three simple shapes are shown in Fig. 8. These calculations found that a simple polished TKTL can provide a fluorescence rejection factor  $>98\%$  - without any Bragg mirrors. This is a remarkable result. This type of link design not only mitigates the thermal resistance issues associated with Bragg mirrors, but also allows direct bonding of the thermal link to the cooling crystal using standard techniques of fusion bonding.



**Figure 8.** Optical rejection with thermal links that do not use Bragg mirrors. For a tapered-kink thermal link design with smooth surfaces (no surface scattering) the rejection is near 99%.



The TDF/UNM team examined another promising compact optical cooler design concept called Diode-pumped Intracavity Solid-state Cryo-Cooler (DISCC) (Fig. 9). This type of cryocooler is based on an intracavity-enhanced absorption scheme using vertical external cavity surface-emitting lasers or VECSEL. (These lasers are also called optically-pumped semiconductor lasers or OPSL.) The diode pump laser light (nominally at 810-900 nm) is delivered by a fiber and focused onto an InGaAs multiple quantum well (MQW) gain chip, designed to operate at  $\lambda=1020$  nm. This chip is grown epitaxially on top of a distributed Bragg reflector (DBR). To control its temperature, the MQW/DBR chip is bonded to a diamond substrate, which is in turn attached to a copper heat sink. The cooling element, a 5-10 mm 5% doped Yb:YLF crystal, is Brewster-cut (to minimize reflection losses) and is held inside a tight “clamshell” chamber using six fused-silica fiber posts. The inside walls of the clamshell are coated with low-emissivity coating, which efficiently absorbs the fluorescence while minimizing the thermal radiation. The laser cavity is completed with a highly reflecting (HR) dielectric mirror. In this design, the thermal link would be attached to the load on the side of the cooling crystal, as shown in Fig. 9. For illustration purposes it is shown as a truncated pyramid of sapphire [JP09].



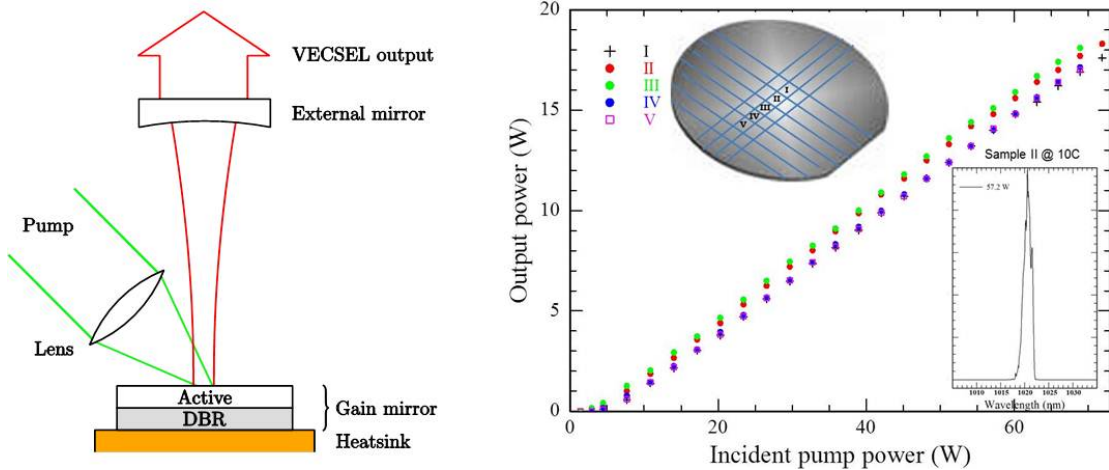
**Figure 9.** Design of a Diode-pumped Intracavity Solid-state Cryo-Cooler (DISCC).

As VECSEL technology is relatively young [OGO10], a high-performance gain chips at specific wavelength are not commercially available. The development of a particular gain chip requires multiple design and epitaxial growth sequences to converge to a working device with optimum characteristics. Dr. Jeffrey Cederberg at Sandia National Laboratories (Albuquerque, NM) grew desired VECSEL chips. After several iterations of growth and characterization, he obtained an operating chip with high quality performance at the designed wavelength of 1020 nm. The design of the VECSEL gain chip, illustrated in Fig. 9, consists of a DBR and a MQW gain region.

The DBR consists of 25 pairs of quarter-wave optical thickness AlAs/GaAs layers. Twelve InGaAs quantum wells (QWs) in a resonant periodic gain structure are placed at adjacent antinodes of the standing wave inside the semiconductor subcavity. The barrier between the QWs, typically GaAs, has been replaced with GaAs<sub>0.97</sub>P<sub>0.03</sub> for strain compensation. The structure is completed by a lattice-matched InGaP window layer, transparent to both the VECSEL emission and pump light, which provides carrier confinement.

Prior to performing intracavity cooling, a simple external cavity setup as shown in Fig. 10 (left) was used to test the VECSEL performance. In this arrangement, the mode size at the VECSEL sample is matched to the roughly 300  $\mu\text{m}$  diameter pump spot created by focusing the light from a fiber-coupled 808 nm pump diode.

For thermal management, which is necessary for high power operation, the DBR is metalized with titanium, gold, and indium and soldered to a 300 $\mu\text{m}$  thick thermal-grade polycrystalline CVD-grown diamond heat spreader, which is metalized in the same way. After the growth, the substrate is removed using a selective wet etch, terminated at the InGaP layer. During operation, the diamond is cooled using a custom water jet impingement technique, where a high velocity liquid jet from a 1-mm diameter nozzle cools the diamond plate directly behind the gain chip.

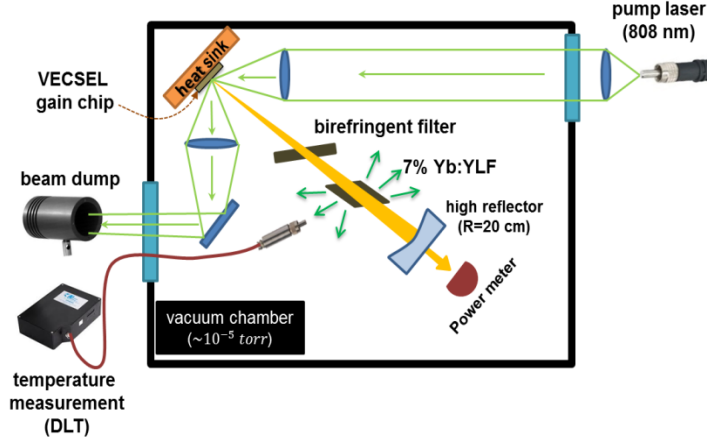


**Figure 10.** (left) Diagram of VECSEL test setup. (right) Data showing the 1020 nm VECSEL chip with record (20%) slope efficiency [ARA12].

As shown in Fig. 10 (right), some of the VECSEL chips operated at 1020 nm with a world record slope efficiencies of 20%. The lasing wavelength and its linewidth can be precisely controlled by inserting spectral selective elements, such as birefringent filters.

Laser cooling experiments were carried out by placing the cooling sample inside of the VECSEL cavity. The cooling sample is a high-purity 7 at. % doped Yb<sup>3+</sup>:YLF crystal cut for E||c pumping with Brewster orientation. A schematic diagram of the intra-cavity laser cooling experiment is depicted in Fig. 11. In order to reduce the

convective heat-load and hence maximize temperature drop in the cooling sample, the experiments are performed in an aluminum vacuum chamber evacuated to a pressure of below  $10^{-5}$  torr using a turbo-molecular pump. The collimated beam from the 808 nm pump laser outside the vacuum chamber is sent through an anti-reflection (AR) coated vacuum-port window.

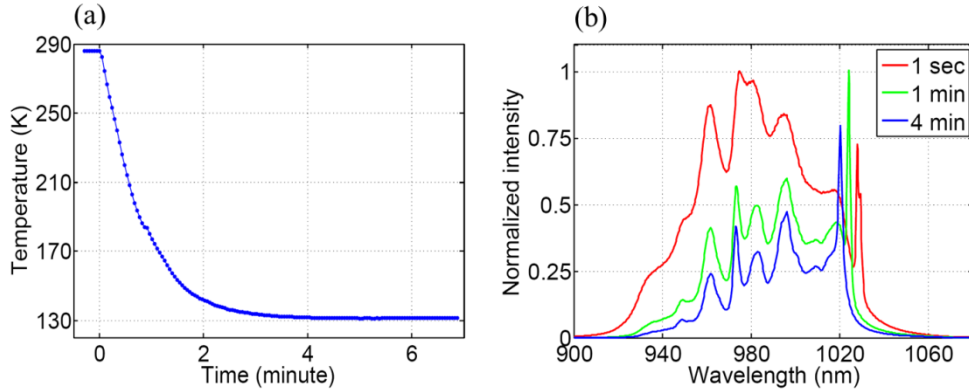


**Figure 11.** Schematic diagram of the VECSEL intracavity laser cooling experiment. High power (75 W) fiber-coupled diode laser at 808 nm is used to pump the VECSEL. A birefringent filter is used to tune the wavelength in the linear cavity with a high reflecting output coupler ( $R=20$  cm). The cooling sample is a Brewster-cut 7% doped Yb:YLF crystal in the E//c orientation. The fluorescence from the sample is collected by a fiber and used to measure the temperature using differential luminescence thermometry method.

The target operation wavelength of the VECSEL is 1020 nm, corresponding to the E4-E5 Stark manifold resonance of the Yb:YLF crystal, which is achieved by rotating the BF around the normal to its face. We note that high absorption loss (in excess of 35% per round-trip) of Yb:YLF crystal at 1020 nm at room temperature prevents the laser operation. Therefore, at room temperature the VECSEL wavelength is tuned to approximately 1030 nm, where the round-trip loss is estimated to be around 9 %, allowing for the operation of the laser. Once the VECSEL starts lasing and cooling the Yb:YLF, we gradually tune the wavelength toward 1020 nm such that the absorbed power in the crystal is maintained despite the decreasing absorption. The thickness of the cooling sample is chosen based on its absorption coefficient to match the optimal coupling loss ( $\sim 4$ -5%). For the optimal cooling wavelength of 1020 nm and at a temperature of 131K, a thickness of approximately 2 mm results in a cavity an estimated roundtrip loss of 6.5 %, close to the optimal coupling.

The dynamics of the cooling is shown in Fig. 12(a), starting from room temperature and reaching 131 K after about 5 minutes. Fluorescence spectra collected during the experiment at different times are compared in Fig. 12(b), showing the change in spectrum used to compute the sample temperature. A clear red-shift of the mean fluorescence wavelength and dramatic narrowing of the emission peaks, together with an overall decrease of the fluorescence counts, are clear signatures of the

sample cooling. A portion of the scattered intracavity laser light, also visible in the spectra, shows the tuning of the VECSEL using the birefringent filter from 1030 nm to 1020 nm as the crystal temperature decreases over time.



**Figure 12.** (a) Yb:YLF crystal temperature as a function of time during cooling experiment. Cooling to 131 K was achieved starting from the room temperature. (b) Luminescence spectra (not corrected for instrument response) at different times; note the scattered intracavity laser light at 1020 nm to 1030 nm.

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## People involved in Phase II research

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Dr. Mansoor Sheik-Bahae (UNM PI)  
Dr. Denis V. Seletskiy  
Dr. Seth D. Melgaard  
Jay V. Alden  
Dr. Alexander R. Albrecht

## Publications resulting in part from Phase II research

“Precise determination of minimum achievable temperature for solid-state optical refrigeration” Seletskiy, D. V., Hehlen, M. P., Melgaard S. D., Epstein R. I., Di Lieto, A., Tonelli, M., Sheik-Bahae, M., *J. Luminescence*, **133**, 5-9, 2013.

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